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Optical properties of a nonlinear $LiInS_2$ crystal

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***Abstract*. The linear and nonlinear optical properties and the radiation resistance of a $LiInS_2$ single crystal are studied. Phase matching conditions are calculated for the first time for a $LiInS_2$ crystal.**

Keywords: *nonlinear crystals, frequency conversion, phase matching.*

Despite a broad transparency spectral range, a rather high nonlinear susceptibility, and an adequate birefringence, the nonlinear semiconductor $LiInS_2$ [1–3] crystals have not attracted the attention of researchers. Being inferior to oxide crystals used in the visible and near-IR ranges in the radiation resistance and to many crystals used in the mid-IR range in the nonlinear susceptibility, these crystals can pretend to the leading position in none of the spectral regions. The difficulty of growing quality samples of adequate sizes is another obstacle, which prevented the estimate of the advantages of $LiInS_2$ crystals over crystals used in the mid-IR range, which can be offered by the presence of lighter Li cations. However, recent technological advances [4, 5] stimulated the study of physical properties of $LiInS_2$ crystals with the aim of determining their potentiality in nonlinear optics.

Biaxial negative $LiInS_2$ crystals belong to the point symmetry group *mm*2. They are nonhygroscopic, have the density 3.5 g cm^{-3}, the melting temperature 880 °C, and the Mohs hardness 3–4 [1]. We studied transparent or slightly yellowish crystals of relatively high quality, of size 4 mm × 4 mm × 4 mm, which were grown by the Bridgman–Stockbarger method. The uncoloured crystals of length 3.6 mm were transparent in the region between 0.4 and 12.5 μm (by the 0.1 level) and in the region between 0.5 and 11 μm (by the 0.5 level) (Fig. 1).

The absorption coefficient in the region of maximum transparency between 1.0 and 8.0 μm was $\alpha \approx 0.1 - 0.25$ cm^{-1} and at the wavelengths of a CO_2 laser between 9.2 and 10.8 μm, $\alpha = 1.1 - 2.3$ cm^{-1}. The short-wavelength boundary of the transparency spectrum for a crystal of length 3.5 mm at the level $\alpha = 200$ cm^{-1} was 330–334 nm at the crystal temperature $T = 80$ K and 342–343 nm at $T = 300$ K for different polarisations of light [4]. The long-wavelength boundary determined for the same level of α was close to 13.2 μm.

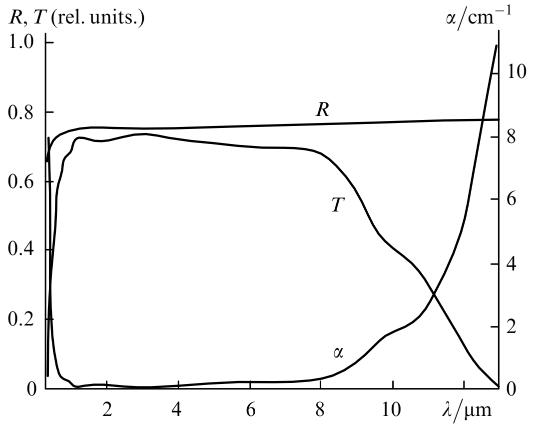

Figure 1. Spectra of Fresnel losses R, transmission T, and the absorption coefficient α of a $LiInS_2$ single crystal of length 3.6 mm.

The dispersion dependences of the refractive index were determined by the least deviation method using prisms and were calculated with the help of the Sellmeyer equation

$$n^2 = A + B/(\lambda^2 - C) - D\lambda^2.$$

with the error $\sim 10^{-3}$. The Sellmaier coefficients are presented in Table 1 for the 0.45–11.5-μm range in a crystal optic coordinate system. No substantial differences between measured refractive indices and their values presented in Ref. [1] were found.

Table 1. Sellmaier coefficients.

Crystal optic axes	A	B	C	D
y	4.418222	0.1254461	0.0657432	0.0028850
x	4.559534	0.1403701	0.069233	0.0028731
z	4.59206	0.1410887	0.069287	0.0030589

The coefficients of the second-order nonlinear susceptibility tensor $d_{31} = 6.2$ pm V^{-1}, $d_{32} = 5.4$ pm V^{-1}, and $d_{33} = 9.8$ pm V^{-1} (measured with an error of ±15 %) proved to be lower approximately by 80 % than their known values

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[1, 5]. They were obtained from comparative measurements of the SHG efficiency by the thing wedge method using a repetitively pulsed CO laser [1]. The coefficients $d_{14} = d_{36}$ of reference ZnGeP$_2$ wedges were assumed equal to 75 pm V^{-1}. Note that the measured and calculated phase-matching angles coincided with an error of no worse than 0.3° (the phase-matching direction is determined by angles θ and φ [6]).

The radiation resistance was measured for leading peaks of the 9.55-μm radiation pulses from a TEA CO$_2$ laser of duration 36 ± 2 ns, which contained no less than 85 % of the total energy of the laser pulse. The damage threshold was no less than 180 MW cm^{-2}.

Analysis of the coefficients of the nonlinear susceptibility tensor showed that the effective nonlinear coefficient for the ssf interaction is nonzero in the xz plane for $\theta < V_z$ (V_z is the angle of inclination to the optical axis z). For the sff and fsf interactions, the effective nonlinear coefficient in nonzero in the xz plane for $\theta > V_z$ and in the xy and yz planes for any θ. The effective nonlinear coefficient is maximum for the second type of interaction in the direction of the y-axis.

The SHG phase-matching properties of a LiInS$_2$ crystal can be conveniently represented with the help of the phase-matching directivity diagram for biaxial crystals [6]. Table 2 presents the numbers of transitions from one projection to another in the diagram [6], for which the phase-matching direction coincides with one of the optical axes. Table 2 also presents the transition wavelengths of the fundamental radiation and the corresponding axes. It follows from these results that phase matching along the x-axis is realised for neither of these wavelengths. The 'loop' type of the transitions in the phase-matching directivity diagram upon variation of the wavelength shows that the crystal should exhibit a particular case of phase matching, which is uncritical to the wavelength – the group phase matching.

Table 2. Transition wavelengths in the SHG phase-matching directivity diagram.

Transition	Interaction type	λ/nm	Axis
00–10	ssf	1573.5	y
10–30	ssf	1731.9	z
30–31	sff	2294.8	y
31–33	sff	2638.3	z
33–31	sff	5104.7	z
31–30	sff	5785.7	y
30–10	ssf	7945.2	z
10–00	ssf	8498.5	y

Fig. 2 shows the dependences of wavelengths λ_1 and λ_2 on the wavelength λ_3 ($\lambda_3^{-1} = \lambda_1^{-1} + \lambda_2^{-1}$), for which the group phase-matching condition is fulfilled in the phase-matching directions of the fsf and sff types in the xy plane. These results show that the frequency conversion (SHG, generation of sum and difference frequencies) of femtosecond pulses can be performed in a broad wavelength range. In particular, LiInS$_2$ crystals are the only known crystals for frequency conversion of femtosecond pulses in the three-micron range. Fig. 3 shows the wavelength-tunable crystal characteristics in the xy plane for different angles φ. These characteristics demonstrate the possible advantage of a LiInS$_2$ crystal over other known crystals for SHG in the three-micron range.

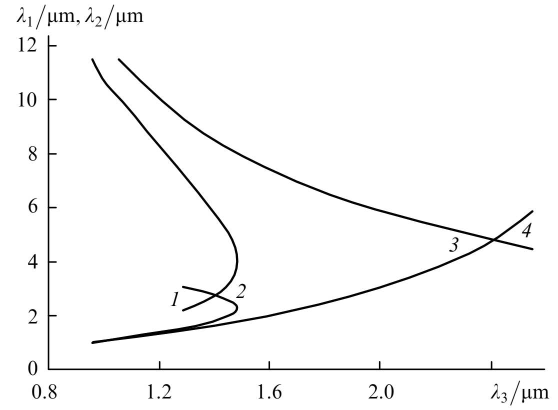

Figure 2. Spectral dependences of the group phase-matching wavelengths for waves s$_1$ and f$_3$ (*1*), s$_2$ and f$_3$ (*2*), f$_1$ and f$_3$ (*3*), and f$_2$ and f$_3$ (*4*) for the sff (*1, 4*) and fsf (*2, 3*) interactions (the subscripts 1, 2, 3 correspond to the number of the wave).

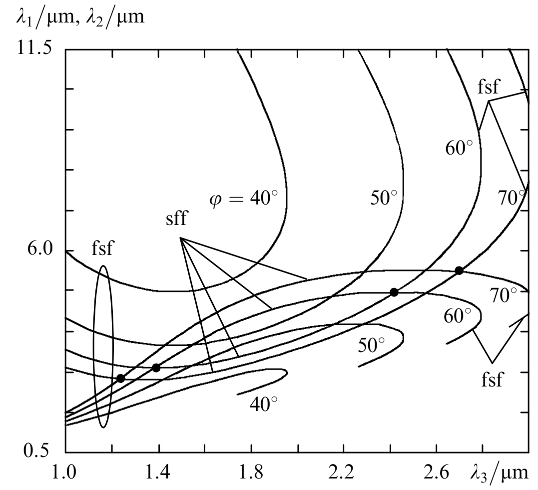

Figure 3. Wavelength-tunable crystal characteristics in the xy plane. Points separate solutions for the sff and fsf interactions.

In conclusion, note that the transparency spectrum, nonlinear properties, and birefringence of LiInS$_2$ single crystals make them promising crystals for frequency conversion of femtosecond pulses. At present, they are the only frequency converters for femtosecond erbium lasers emitting in the three-micron range. The phase-matching conditions were calculated using the LID-SHG applied program set (www.bmstu.ru/~lid).

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